NONLINEAR PROPERTIES IN LANGASITE ISOMORPHS FOR ADVANCED FREQUENCY CONTROL DEVICES AND CLOCKS

Yoonkee Kim

U.S. Army Communications-Electronics Command, Research Development & Engineering Center

Fort Monmouth, New Jersey, 07703

Abstract

Precision oscillators are an enabling technology for precision-guided munitions, slow-moving target detection and identification from moving platforms, and rapid signal acquisition in jamming environments. Quartz has been exclusively used for resonators, which are essential components in such oscillators. Most recently, langasite and its isomorphs (LGX) have been advanced as potential substitutes for quartz, owing to their extremely high quality (Q) factors. A higher Q value, by at least 2.5 times, than that of quartz has been reported. A high Q value translates into a potentially greater stability. In order to make such materials practical, the environmental sensitivities must be addressed. In this paper, some of the nonlinear properties we have characterized over the past years are summarized.

1. INTRODUCTION

In recent years, langasite isomorphs, such as langasite ($La_3Ga_5SiO_{14}$ or LGS), langanite ($La_3Ga_{5.5}Nb_{0.5}O_{14}$ or LGN), and langatate ($La_3Ga_{5.5}Ta_{0.5}O_{14}$ or LGT), have emerged as new materials to replace quartz in advanced frequency control devices and clocks. Langasite is a synthetically grown material originally developed for host crystals for lasers in Russia in the 1980s. It was soon found that its optical properties did not meet expectations. However, since it is piezoelectric, research has been focused on utilizing its piezoelectricity for frequency control applications. In the 1990s, it was discovered that langasite isomorph has a higher Q value, by at least 2.5 times, and a higher piezoelectric coupling than quartz (Smythe et al., 2000). The Q values for LGN and LGT are particularly impressive in comparison to quartz as reflected in Q·frequency ($Q{\cdot}f$) products as large as 29×10^6, where f is in MHz, for unplated Y-cut plano-convex LGN (GTS, 2000) and LGT (Smythe et al., 2000) resonators driven by thickness-field-excitation. To put these values in perspective, consider that the upper bound $Q{\cdot}f$ for AT-cut natural quartz resonators is 15×10^6 (Warner, 1960). Temperature-compensated orientations also exist for thickness-shear waves (c mode). Presently, four-inch langasite wafers are commercially produced for manufacturing surface-acoustic-wave (SAW) IF-filters for W-CDMA base stations since the langasite filters can provide steeper cutoff skirts of the band-pass filters than the quartz filters. With the high Q value and the existence of temperature compensated cuts for thickness-shear waves (c-mode), langasite isomorph may also produce better precision bulk-acoustic-wave (BAW) resonators for frequency standards and clocks than quartz. Furthermore, by having no phase transition temperature up to its melting point at 1470 °C, langasite isomorph may be used as the material of choice for sensors operated under a very high temperature condition (Thiele et al., 2005).

In order to use langasite isomorph for BAW resonators, the environmental sensitivities must be addressed. Knowledge of nonlinear properties is critical for the design of resonators. Some nonlinear properties are force-frequency, acceleration-frequency, resonance amplitude-frequency, intermodulation, mode coupling-activity dips, dynamic thermal-frequency, and film stress-frequency (Ballato et al., 1977). Other than the first-order material constants (stiffness, piezoelectric, and dielectric) and quasi-static frequency-temperature characteristics, nonlinear properties are not well characterized for this material. To address this issue, for the last few years we have been investigating singly-rotated Y-cut LGX resonators for force-frequency effect, acceleration-sensitivity, amplitude-frequency effect, and thermal transient effect (Kim and Ballato, 2003; Kim, 2003; Kim, 2004). Our investigation results are summarized in this paper. Detailed description of the measurement methodologies for characterizing nonlinear properties can be found in our publications and are not duplicated here.

The langasite isomorphs and test resonators that were characterized have been fabricated under an Army contract (N66001-97-C-8634) of which objective was to determine the material constants of langasite isomorphs (Smythe et al., 2000). Table 1 lists the parameters of the LGN and the LGT resonators investigated here. The 3rd, 5th, and 7th overtones (OT) of the slow-shear mode (c-mode) were investigated since the spectrum of each mode was clean in the vicinity of the resonance. Fig. 1 shows an LGT boule recently grown at the Univ. of Central Florida and some LGT test resonators similar to the resonators investigated for the nonlinear properties.

2. FORCE-FREQUENCY EFFECT

It has been determined that the sensitivity of the resonator frequency to shock and vibration could be reduced

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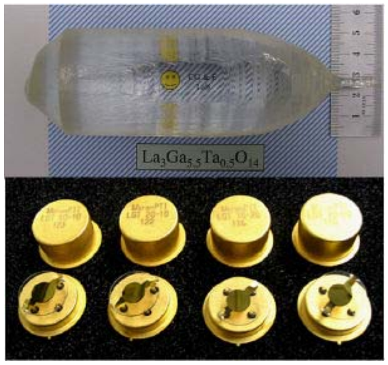

Fig. 1. Langatate boule (photo courtesy of the Univ. Central Florida) and typical resonators for testing.

Table 1. Resonator parameters.

	Y-cut LGN	Y-cut LGT
Crystal disk	Plano-convex, 14 mm Ø	
Thickness [mm]	0.71	0.64
diopter	3	2
Electrodes	6.35 mm Ø gold with chrome adhesion layer	
Slow-shear velocity [m/s]	2859.36	2600.32
3rd OT [MHz]	6.047	6.149
5th OT [MHz]	10.082	10.254
7th OT [MHz]	14.102	14.347

by selecting certain mounting support orientations of the crystal resonator blank. The phenomenon of frequency changes related to the stress applied to the resonator is referred to as the force-frequency effect. The frequency changes are due primarily to the nonlinear (3rd order non-Hookean) elastic constants and secondly to the static deformation of the crystal lattice. The knowledge of the coefficients applies to the design of mounting supports for resonators so that the mounting stress effects may be minimized. The force-frequency effects also contribute to long-term aging.

Fig. 2 shows the definition of the azimuthal angle ψ between the direction of applied force and the crystallographic axis of a singly-rotated cut. The force-frequency effects produced in a circular crystal plate acted upon by a diametric in-plane force at the angle ψ is characterized by means of a force-frequency coefficient K_f, which is defined by (Ratajski, 1968)

$$K_f = \frac{\Delta f}{f} \cdot \frac{\text{(Diameter)(Thickness)}}{\text{(Force)(Acoustic velocity/2)}} \tag{1}$$

where $\Delta f/f$ is the normalized frequency change due to the applied force. A smaller K_f is desirable for precision clock and frequency control applications, whereas a larger K_f is more suitable for transducer applications. Another important factor is a coefficient of planar-stress, which is defined by an integral of K_f over the perimeter (Ballato et al., 1977), as follows

$$\langle K_f \rangle = \frac{1}{\pi} \int_0^{\pi} K_f(\psi) d\psi \, . \tag{2}$$

It represents the superposition of a continuous distribution of periphery stresses. Crystal cuts in the vicinity of $<K_f> = 0$ are especially important. They are referred to as stress-compensated (SC) cuts. An ideal SC cut resonator is insensitive to planar stress such as electrode film stress at a certain temperature.

Fig. 3 contains the measured K_f values of Y-cut LGN, Y-cut LGT, and AT-cut and SC-cut quartz. Zero crossings of K_f exist for both materials. Note that LGX has a substantially smaller K_f than quartz over the entire range of Ψ. Table 2 lists some notable data points, *i.e.* the azimuthal angles for $K_f = 0$ and K_f = extrema, and extrema K_f and $<K_f>$ values. The positive (negative) extrema of K_f of the LGN and the LGT are 7 and 10 (5 and 8) times smaller than those of AT-cut quartz, respectively. The small K_f values of the LGN and the LGT reflect the smaller $<K_f>$ values by 18 and 25 times than that of AT-cut quartz, respectively. Exact numbers may deviate slightly from the numbers shown due to measurement error. The Y-cut LGN and the Y-cut LGT show very similar shapes with the same zero crossing angles at ~52° and ~128° except that the overall magnitudes of K_f of the LGN are larger by ~44% than those of the LGT.

Vibrational force contributes frequency excursions and K_f correlates diametric (static) force to frequency excursions. Questions arise, naturally, on how the small K_f value guarantees low acceleration sensitivity. One may argue that K_f reveals only the simple effects related to the

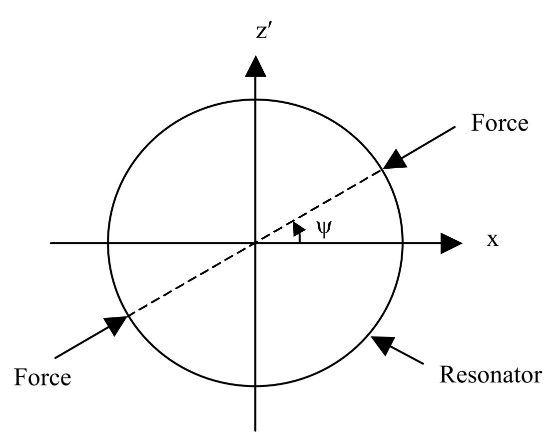

Fig. 2. Definition of force application and of angle Ψ.

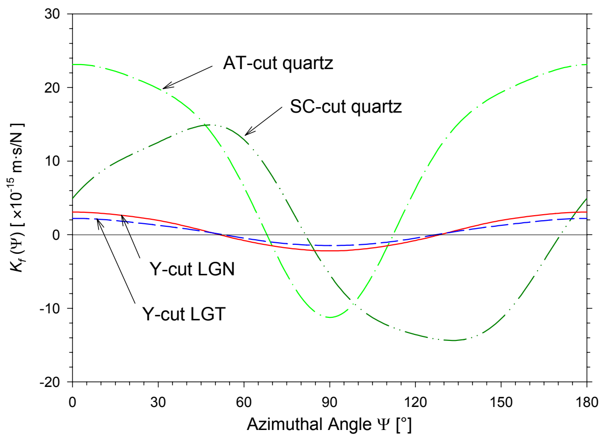

Fig. 3. Comparison of K_f between LGX and quartz.

Table 2. Comparison of K_f and $< K_f >$ between LGX and quartz. Dimension of K_f and $< K_f >$ is 10^{-15} m·s/N.

Material	Ψ [°] for K_f=0	Ψ [°] for K_f= extrema	Extrema K_f	$< K_f >$
Y-cut LGN	51 129	0 90	3.1 –2.2	0.52
Y-cut LGT	53 127	0 90	2.2 –1.5	0.38
AT-cut quartz	68 112	0 90	23 –11.3	9.4 (exp.) 10.0(theo.)
SC-cut quartz	81 171	48 133	15 –13.4	0

diametric force, but the vibrational force is more complicated, including combinations of all directions, bending, twisting, and twitching. The nature of such complication can easily be explained by the fact that AT-cut quartz sometimes shows lower acceleration sensitivity than SC-cut quartz even though the maximum K_f value of AT-cut is larger than that of SC-cut. Force-frequency effects for general directions may be readily quantified by investigating K_f for many different cuts of the material. Previous investigations on quartz for several rotated cuts reveal that the maximum magnitudes of K_f fall within the same order of the magnitudes (Ballato et al., 1977). It is highly probable that LGX also behaves similarly to quartz with respect to K_f. Among myriads of cuts including doubly-rotated cuts, SC-cuts may exist for LGX too. With SC-cuts found, a smaller K_f implies that LGX would have lower film stress-frequency and thermal transient effects than quartz. Combined with the high Q of the material, the material is promising for yielding lower acceleration sensitivity and lower noise resonators than quartz.

3. ACCELERATION SENSITIVITY

Acceleration sensitivity is one of the important nonlinear properties of a resonator, especially when it is subjected to undergo vibration. A resonator with high acceleration sensitivity can increase the phase noise floor severely to dominate any other noise sources. The acceleration sensitivities range from a few parts in 10^9/g for typical quartz resonators to the low parts in 10^{10}/g for a state-of-the-art SC-cut resonator mounted on a specially designed package to passively achieve a low acceleration sensitivity. With an active cancellation technique, the level can be lowered by an order to ~10^{-11}/g.

The acceleration sensitivities of two-point mounted langasite resonators were measured to be 2~6×10^{-10}/g for the Y-cut LGT and 6~10×10^{-10}/g for the Y-cut LGN. Two-point mounted resonators usually have poor acceleration sensitivities due to an extra stress component from the mount bending along the direction connecting mounting points. Nevertheless, the LGT shows low values of acceleration sensitivity that is comparable to state-of-the-art quartz, implying that, while being mounted on a similar package constructed for the low acceleration sensitivity quartz, there is a potential for yielding low acceleration sensitivity of 10^{-11}/g level passively and super-low ~10^{-12} /g with an active cancellation, assuming the same principle employed for quartz applies.

4. AMPLITUDE-FREQUENCY EFFECT

One of the important non-linear effects needed to be known is the amplitude-frequency effect where resonant frequency changes as a function of drive current due to nonlinear elastic constants of a resonator crystal (Hammond et al., 1963; Gagnepain and Besson, 1975). Theoretical analyses of the effect are found in Refs. (Tiersten, 1975; Tiersten, 1976). In quartz resonators, the amplitude-frequency effect may be adjusted by the design of the resonator, such as crystal cut, contouring, and overtone (Kusters, 1981; Filler, 1985). A lower amplitude-frequency effect implies more insensitivity to current fluctuations and slow current drift in an oscillator circuit, contributing to better short-term stability and lower long-term aging respectively (Filler, 1985).

Fig. 4 shows the measured magnitude and phase of the 7th OT of an LGN resonator as a function of the input voltage to an amplifier of which output drives a test resonator as a part of a load consisting of a simple resistor network. The current flow through the resonator, by applying the input voltage, was directly measured with a current probe. As the current increases, the magnitude curves bend toward lower frequencies. The phase curves also shift downward correspondingly. This downward

shift (*i.e.*, so-called a soft spring effect) is also observable at BT-cut quartz (Ballato, 1983). The different shapes in the magnitudes and the phases imply the motional impedance of the resonator also changes. Note that an abrupt current jump occurs at the driving voltage of 200 mV. When such a jump occurs, the phase becomes steep and obviously the resonant frequency deviates from the typical trend in the frequency bending.

With such frequency shifts measured as a function of current, Fig. 5 plots typical frequency-versus-current data of the 3rd, 5th, and 7th OT of the LGT with down and up sweep, along with typical curves of AT-cut and SC-cut quartz. The two sweeps produced very similar curves for each OT. The data set of each OT and each sweep was fitted with a quadratic curve to obtain a drive sensitivity coefficient D defined by (Filler, 1985)

$$\Delta F_{A-F} / F = D \cdot i^2 . \tag{3}$$

For each OT, the average value of the D's of the two sweeps was taken. For the fundamental modes, instead of showing a quadratic characteristic, the $\Delta F_{A\text{-}F}$ data points were scattered within ± 0.5 ppm, and Q was 2~3×10^5 for the fundamental modes (at least one order higher for the OT's for both materials). Thus, it was not possible to obtain the D for the fundamental modes.

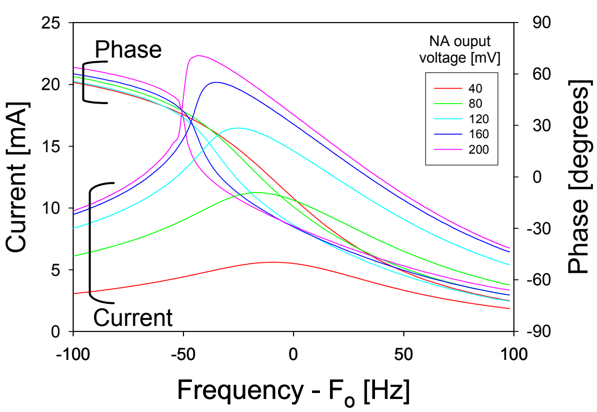

Fig. 4. Typical measured data and curve as a function of network analyzer output voltage- (a) Current magnitude, (b) Phase.

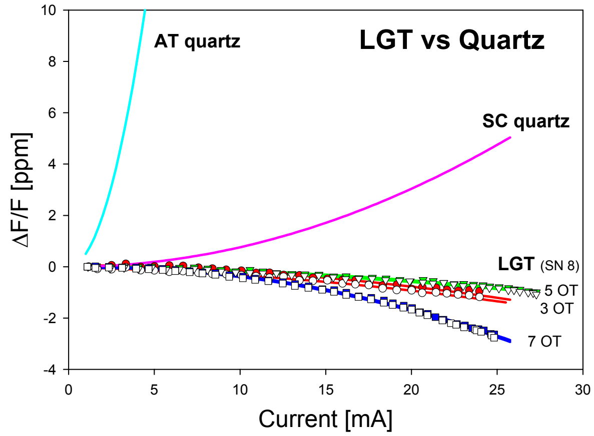

Fig. 5. Mode frequency changes versus currents at resonant frequencies of (a) LGN, (b) LGT. Curve-fit to measured data of down and up sweep frequencies are shown as solid lines.

Three of each LGN and LGT resonator were measured and the results are listed in Table 3. Obtaining the D's for the 7th OT of SN10 was not possible either, due to the anharmonic mode interference. The D's of the 3rd OT's and the 5th OT's are in the vicinity of –2 ppb̃mA^2 for both resonators. The 5th OT's have slightly lower D's than the 3rd OT's. The 7th OT's have higher D's than the other OT's. The lowest drive sensitivity is –1.2 ppb̃mA^2 on the 5th OT of the LGT. Overall, the LGT has smaller D's than the LGN. Knowing that the coefficient depends

Table 3. Drive sensitivity coefficient D of LGN and LGT. (SN is an arbitrary serial number and Avg is the average value of 3 resonators.) Obtaining D for 7th OT of SN10 was not possible due to anharmonic mode interference.

LGN			
SN	3rd OT	5th OT	7th OT
6	-3.85	-2.65	-6.85
7	-3.30	-2.94	-8.66
10	-3.15	-2.66	–
Avg	-3.43	-2.75	-7.76
LGT			
SN	3rd OT	5th OT	7th OT
8	-1.65	-1.06	-4.91
26	-1.50	-1.25	-5.68
28	-1.74	-1.19	-5.47
Avg	-1.63	-1.17	-5.35

on not only the material constants but also the design parameters of the resonator such as its crystal cut, contouring, and overtone (Filler, 1985), we cannot identify the cause of the difference. More measurements with various design parameters will clarify whether it is due to the intrinsic material properties or the design parameters.

The drive sensitivity of LGT is compared with those of quartz (Drive sensitivity: –1.2 ppb/mA^2 for LGT, 504 ppb/mA^2 for AT-cut quartz, 7.6 ppb/mA^2 for SC-cut quartz) (Gagnepain, 1975). As shown in Fig. 5, it is easily seen that langasite isomorphs have much smaller amplitude-frequency effects than those of quartz. Having a smaller amplitude-frequency effect implies the possibility of producing a better short-term stability and a lower long-term aging property.

5. THERMAL TRANSIENT EFFECT

When a piezoelectric resonator undergoes a rapid temperature change, the resonant frequency deviates from that of a static temperature case. Such a temperature change creates a thermal gradient inside of the resonator to cause a localized stress change in the crystal along with changes in material constants such as elastic modulus and density (Holland, 1974). In addition, due to different thermal expansion coefficients between the crystal and its mounting supports, the supports apply an in-plane diametric force to the crystal (Janiaud et al., 1981). The combination of these factors causes the frequency deviation.

The thermal transient effect on a resonator frequency may be quantified with the dynamic thermal sensitivity coefficient $\tilde{a}$, as defined in Ref. (Janiaud et al., 1981; Koechler et al., 1977; Young et al., 1978),

$$\frac{\Delta f(t)}{f} = \tilde{a}\frac{dT(t)}{dt} \tag{4}$$

where Δf is the frequency shift with respect to the static thermal frequency, T is the temperature, and t is the time. To obtain $\tilde{a}$, it is a usual practice that the Δf is measured at a turnover (TO) frequency (Kusters, 1976). Applying simple algebra to Eq. (4) yields the following equation,

$$\tilde{a} = \left(\frac{\Delta f_1(t)}{f} - \frac{\Delta f_2(t)}{f}\right) \Big/ \left(\frac{dT_1(t)}{dt} - \frac{dT_2(t)}{dt}\right) \tag{5}$$

where 1 represents a temperature increase and 2 represents a temperature decrease subsequent to the increase. Both temperature increase and decrease pass though a TO frequency to obtain $\Delta f = \Delta f_1 - \Delta f_2$ at the TO frequency. The temperature increase and decrease rates are maintained as linear ramps to produce constant derivatives with respect to time.

Fig. 6 shows typical normalized static F-T data of a Y-cut LGN and a Y-cut LGT from the fundamental mode to

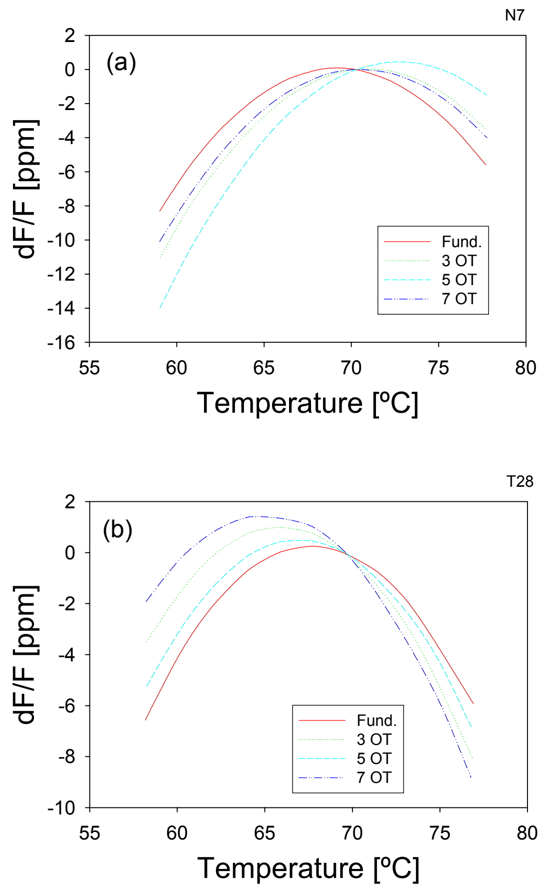

Fig. 6. Parabolic static frequency-temperature curves: (a) Y-cut LGN, (b) Y-cut LGT.

the 7th OT. The F-T curves are parabolic. The TO temperatures vary among the modes and the units with the range from 55 °C to 75 °C. Each mode frequency is referenced to the mode frequency at room temperature, as denoted by dF.

The Y-cut LGT shows that the Δf has a linear relationship to the ramping rate, resulting in the constant values of the $\tilde{a}$, *i.e.,* independent of the rate. In contrast, the Y-cut LGN does not show such a linear relationship and the sign of the $\tilde{a}$ changes from negative to positive for the 3rd and 5th OT depending on the rate. The LGN shows smaller magnitudes of the ã than the LGT for the fast ramping rates, but larger magnitudes for the slow rates. The OT's have smaller values than the fundamental modes. For the 7th OT, only three resonators were measurable because of activity dips. The mean values of the $\tilde{a}$ shown in Fig. 7 are provided in Table 4. The two-point-mount shows smaller values by ~33 % than the four-point-mount. Due to the sign change, the ranges are given for the LGN instead of the mean values.

Now, compare the Y-cut LGX to an AT-cut quartz. The measured $\tilde{a}$ values for 5 MHz 5th OT AT-Cut quartz

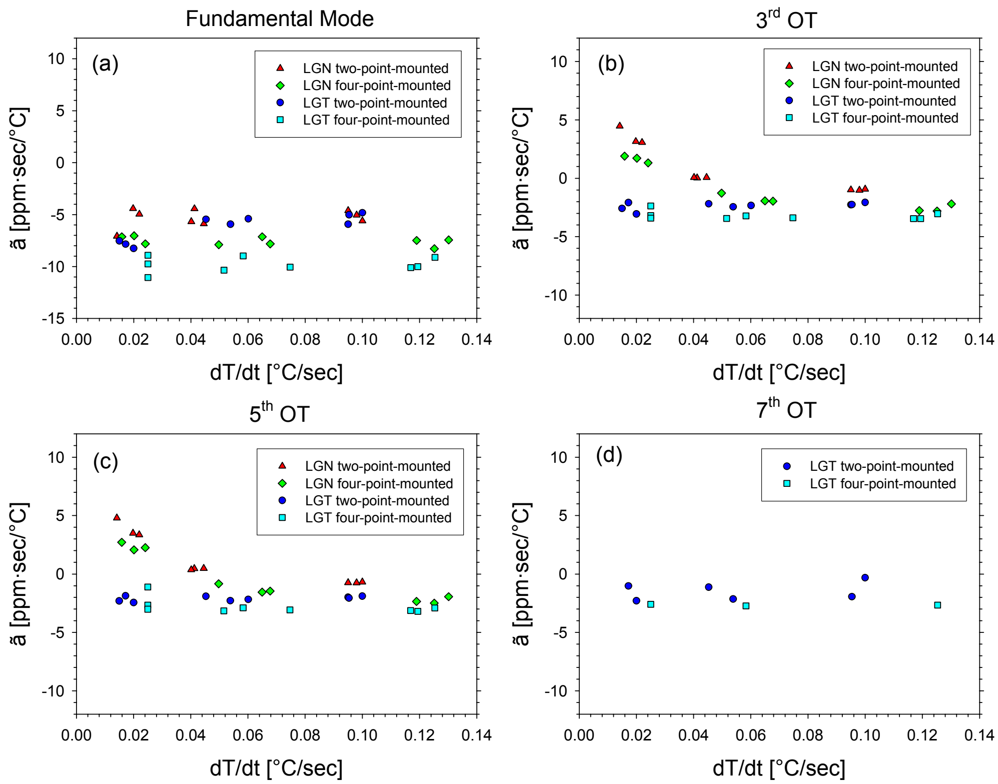

Fig. 7. Measured values of ã for two- and four-point-mounted Y-cut LGN and LGT: (a) fundamental mode, (b) 3rd OT, (c) 5th OT, and (d) 7th OT.

Table 4. Mean values of ã. The dimension of ã is [ppm·s/°C].

Material		Y-cut LGN		Y-cut LGT	
Mount		Two	Four	Two	Four
Mode	Fund.	–5.3	–7.6	–6.2	–9.8
	3rd OT	–1 ~4.5	–2.8 ~1.9	–2.4	–3.2
	5th OT	–0.8 ~4.8	–2.4 ~2.7	–2.1	–2.8
	7th OT	-	-	–1.5	–2.7

resonators were reported as –15.3 ~ –13 ppm·s/°C (Janiaud et al., 1981; Young et al., 1978; Kusters, 1976). The *ã* value of the 10 MHz 5th OT two-point-mounted LGT is only 15% of that of the AT-cut quartz. This is partly because the LGT is less susceptible to the dynamic stress changes from the mounting supports due to different thermal expansion coefficients between the crystal and the mounting supports. This result also agrees with the aforementioned measurements on the same LGX materials, which have lower force-frequency effects than AT-cut quartz. The thermal transient effect also contributes to its short-term stability.

6. CONCLUSIONS

Y-cut langasite isomorphs show much smaller non-linear properties of force-frequency, amplitude-frequency, and thermal transient effects in comparison to quartz. Combined with its high Q value, langasite isomorph has a potential to substitute quartz in advanced frequency control and timing applications. Especially, the force-frequency effect is smaller by almost an order of magnitude, implying that, while being mounted on a similar package constructed for the low acceleration sensitivity quartz, there is a potential for yielding low acceleration sensitivity of 10^{-11}/g level passively and super-low ~10^{-12} /g with an active cancellation, assuming the same principle employed for quartz applies.

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